

#### IV. INTERACTION HAMILTONIAN -- COUPLING FIELDS AND CHARGES:<sup>23</sup>

To build a complete quantum picture of the interaction of matter and radiation our first and most critical task is to construct a *reliable* Lagrangian-Hamiltonian formulation of the problem. In this treatment, we will confine ourselves to a **nonrelativistic view** which, fortunately, is adequate for most circumstances. We start with a representation of the Lorentz force for a single charged particle -- viz.

$$\vec{f} = q \left\{ \vec{E} + \vec{v} \times \vec{B} \right\} \quad [IV-1a]$$

or in terms of the electromagnetic potentials

$$\vec{f} = q \left[ -\vec{\nabla} \phi - \frac{1}{c} \frac{d\vec{A}}{dt} + \vec{v} \times \left( \vec{\nabla} \times \vec{A} \right) \right] \quad [IV-1b]$$

Let us now write the **total time derivative** of a component of the vector potential

$$\frac{dA_r}{dt} = \frac{A_r}{r} \frac{dr}{dt} + \frac{A_r}{t} = \left( \vec{v} \cdot \vec{\nabla} \right) A_r + \frac{A_r}{t} \quad [IV-2]$$

so that

$$\vec{v} \times \left( \vec{\nabla} \times \vec{A} \right) = \vec{\nabla} \left( \vec{v} \cdot \vec{A} \right) - \left( \vec{v} \cdot \vec{\nabla} \right) \vec{A} = \vec{\nabla} \left( \vec{v} \cdot \vec{A} \right) - \frac{d\vec{A}}{dt} + \frac{\vec{A}}{t} \quad [IV-3]$$

Therefore, we may write the Lorentz force as

$$\vec{f} = q \left[ -\vec{\nabla} \phi - \frac{1}{c} \frac{d\vec{A}}{dt} + \vec{v} \times \left( \vec{\nabla} \times \vec{A} \right) \right] \quad [IV-4]$$

<sup>23</sup> Much of what follows draws heavily on material in Chapter 5 of Rodney Loudon's *The Quantum Theory of Light* (second edition), Oxford (1983). Marlan O. Scully and M. Suhail Zubairy in Section 5.1 of *Quantum Optics*, Cambridge (1997) have a slightly different, but quite insightful treatment of the subject of the atom-field interaction Hamiltonian.

We may also write 
$$-A = \frac{1}{v} \left[ -v A \right]$$

so that Equation [ IV-4] becomes

$$\begin{aligned} f &= -\frac{d}{dt} \left\{ q \left[ -v A \right] \right\} + \frac{d}{dt} \frac{1}{v} \left\{ q \left[ -v A \right] \right\} \\ &= m \frac{d}{dt} v = m \frac{d}{dt} \frac{1}{v} \frac{1}{2} v^2 = \frac{d}{dt} \frac{1}{v} \frac{1}{2} m v^2 \end{aligned} \quad [ \text{IV-5} ]$$

which may, in turn, be written

$$\frac{d}{dt} \frac{1}{v} \frac{1}{2} m v^2 - q \left( -v A \right) - \frac{d}{dt} \frac{1}{v} \frac{1}{2} m v^2 - q \left( -v A \right) = 0 . \quad [ \text{IV-6} ]$$

This last equation may now be compared to the **Lagrangian equation of motion** -- *i.e.*

$$\frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{r}}} - \frac{\partial \mathcal{L}}{\partial \mathbf{r}} = 0 \quad [ \text{IV-7a} ]$$

where, in general,

$$\mathcal{L}(\mathbf{r}, \dot{\mathbf{r}}) = [\text{Kinetic Energy}] - [\text{Potential Energy}] = \mathcal{T}(\mathbf{r}, \dot{\mathbf{r}}) - \mathcal{U}(\mathbf{r}, \dot{\mathbf{r}}) . \quad [ \text{IV-7b} ]$$

Therefore, we identify

$$\mathcal{L}(\mathbf{r}, \dot{\mathbf{r}}) = \frac{1}{2} m \dot{\mathbf{r}}^2 - q \left( -\dot{\mathbf{r}} \cdot \mathbf{A} \right) \quad [ \text{IV-8} ]$$

as the Lagrangian for a single charged particle. We may write

$$\frac{d}{dt} \mathcal{L}(\mathbf{r}, \dot{\mathbf{r}}) = \frac{\partial \mathcal{L}}{\partial \mathbf{r}} \cdot \dot{\mathbf{r}} + \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{r}}} \cdot \ddot{\mathbf{r}} \quad [ \text{IV-9a} ]$$

which, in light of Equation [ IV-8 ], becomes

$$\begin{aligned} \frac{d}{dt} \mathcal{L}(\vec{r}, \vec{v}) &= \frac{d}{dt} \frac{1}{v} \mathcal{L}(\vec{r}, \vec{v}) v + \frac{1}{v} \mathcal{L}(\vec{r}, \vec{v}) \dot{v} \\ &= \frac{d}{dt} v \frac{1}{v} \mathcal{L}(\vec{r}, \vec{v}) \end{aligned} \quad [ \text{IV-9b} ]$$

Therefore,

$$v \frac{1}{v} \mathcal{L}(\vec{r}, \vec{v}) - \mathcal{L}(\vec{r}, \vec{v}) = \mathbf{A} \text{ constant of motion} \quad [ \text{IV-9c} ]$$

and, using the canonically conjugate momenta associated Equation [ IV-8 ] -- *i.e.*

$$\mathcal{P} = \frac{1}{v} \mathcal{L}(\vec{r}, \vec{v}) = m v + q A \quad --, \quad [ \text{IV-10} ]$$

we see that Equation [ IV-9c ] can be written

$$\begin{aligned} \frac{1}{m} [\mathcal{P} - q A] \mathcal{P} - \frac{1}{2m} [\mathcal{P} - q A] [\mathcal{P} - q A] + q - \frac{q}{m} A [\mathcal{P} - q A] \\ = \mathbf{A} \text{ constant of motion} \end{aligned} \quad [ \text{IV-11a} ]$$

or finally

$$q + \frac{1}{2m} \left[ \vec{\mathcal{P}} - q \vec{A} \right]^2 = \mathbf{A} \text{ constant of motion} \quad [ \text{IV-11b} ]$$

We are now in a position to set forth the **nonrelativistic Hamiltonian of a single charged particle** -- *viz.*

$$\mathcal{H} = q + \frac{1}{2m} \left[ \vec{\mathcal{P}} - q \vec{A} \right]^2 \quad [ \text{IV-12} ]$$

with the canonical conjugate variables given by

$$\dot{\mathbf{r}} = \frac{\mathcal{H}}{\mathcal{P}} = \frac{1}{m} [\mathcal{P} - q\mathbf{A}] = \mathbf{v} \quad [\text{IV-13a}]$$

$$\dot{\mathcal{P}} = -\frac{\mathcal{H}}{\mathbf{r}} = -q\frac{1}{\mathbf{r}} + \frac{1}{m} [\mathcal{P} - q\mathbf{A}] \frac{1}{\mathbf{r}} [q\mathbf{A}] \quad [\text{IV-13b}]$$

In principal, we are **done**, since we may now write the complete Hamiltonian for a many particle material system as

$$\mathcal{H} = \sum_i \mathcal{H}_i = \sum_i q_i + \frac{1}{2m_i} [\vec{\mathcal{P}}_i - q\vec{\mathbf{A}}]^2 \quad [\text{IV-14}]$$

Unfortunately, this form of the Hamiltonian is not the most useful in optical physics since it is expressed in terms of the vector potential and needs to be evaluated at all charge positions. Most annoyingly, it does not yield an interaction term in the form used earlier - *i.e.* Equation [ III-3a ] in the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory*.

### THE MULTIPOLE EXPANSION OF THE CLASSICAL HAMILTONIAN:

Before we manipulate the Hamiltonian further, we digress a bit to get a better fix on what we are really look for. From the most basic notions of electrostatics, the energy of interaction **with an external transverse field** should be expressible as the “energy of assembly”<sup>24</sup> -- viz.

$$- \sum_i q_i \int_{\vec{\mathbf{r}}_{\text{ref}}}^{\vec{\mathbf{r}}_i} \vec{\mathbf{E}}_{\text{T}}(\vec{\mathbf{r}}) d\vec{\mathbf{r}} = - \sum_i q_i \int_{\vec{\mathbf{r}}_{\text{ref}}}^{\vec{\mathbf{r}}=0} \vec{\mathbf{E}}_{\text{T}}(\vec{\mathbf{r}}) d\vec{\mathbf{r}} + \sum_i q_i \int_{\vec{\mathbf{r}}=0}^{\vec{\mathbf{r}}_i} \vec{\mathbf{E}}_{\text{T}}(\vec{\mathbf{r}}) d\vec{\mathbf{r}} \quad [\text{IV-15a}]$$

<sup>24</sup> This is the work required to assemble the atom in the external field and of course ignore all inter-particle electrostatic interactions

which becomes for a neutral *atom* with charge centered at  $\vec{r} = 0$

$$U_{\text{int}} = \sum_i q_i \int_{\vec{r}=0}^{\vec{r}_i} \vec{E}_T(\vec{r}) d\vec{r} \quad [\text{IV-15b}]$$

Electric fields of interest vary only slightly over an *atom* so that we should be able to expand the external field in a rapidly converging series as follows

$$\vec{E}_T(\vec{r}) = \vec{E}_T(0) + \left( \vec{r} \cdot \nabla \right) \vec{E}_T(\vec{r}) \Big|_{\vec{r}=0} + \frac{1}{2!} \left( \vec{r} \cdot \nabla \right)^2 \vec{E}_T(\vec{r}) \Big|_{\vec{r}=0} + \dots \quad [\text{IV-16}]$$

Substituting this expansion into Equation [ IV-15b ] and integrating we obtain

$$U_{\text{int}} = -e \sum_i \vec{r}_i \left[ 1 + \frac{1}{2!} \left( \vec{r}_i \cdot \nabla \right) + \frac{1}{3!} \left( \vec{r}_i \cdot \nabla \right)^2 + \dots \right] \vec{E}_T(\vec{r}) \Big|_{\vec{r}=0} \quad [\text{IV-17a}]$$

Formally, we may cast this expansion for the interaction energy in the form

$$U_{\text{int}} = -e \sum_i \vec{r}_i \frac{\exp(\vec{r}_i \cdot \nabla) - 1}{\vec{r}_i \cdot \nabla} \vec{E}_T(\vec{r}) \Big|_{\vec{r}=0} \quad [\text{IV-17b}]$$

In the continuum picture, the interaction energy should be expressible in the form

$$U_{\text{int}} = \int \vec{E}_T(\vec{r}) \cdot \vec{P}(\vec{r}) dV \quad [\text{IV-18}]$$

where  $\vec{P}(\vec{r})$  is the polarization density. It may be shown quite easily that Equations [ IV-18 ] and [ IV-17 ] are equivalent if the polarization density is expressed in the following

***multipole expansion***

$$\vec{\mathbf{P}}(\vec{\mathbf{r}}) = -e \sum_i^{\text{electrons}} \left\{ \vec{\mathbf{r}} - 1/2! \vec{\mathbf{r}}(\vec{\mathbf{r}} - \vec{\mathbf{r}}_i) + 1/3! \vec{\mathbf{r}}(\vec{\mathbf{r}} - \vec{\mathbf{r}}_i)^2 + \dots \right\} (\vec{\mathbf{r}} - \vec{\mathbf{r}}_i) \quad [\text{IV-19}]$$

With this background, we return to a consideration of the Hamiltonian in Equation [ IV-14 ]. In what follows, we show that this Hamiltonian is transformed into a form which consistent with Equation [ IV-17b ] if we make an appropriate **gauge transformation** of the fields. In general, the gauge transformation

$$\begin{aligned} \vec{\mathbf{A}}(\vec{\mathbf{r}}, t) &= \vec{\mathbf{A}}(\vec{\mathbf{r}}, t) - \vec{\nabla} \chi(\vec{\mathbf{r}}, t) \\ \chi(\vec{\mathbf{r}}, t) &= \chi(\vec{\mathbf{r}}, t) + \frac{1}{t} \chi(\vec{\mathbf{r}}, t) \end{aligned} \quad [\text{IV-20}]$$

where  $\chi(\vec{\mathbf{r}}, t)$  is an arbitrary scalar gauge function, leaves the electric and magnetic fields unchanged. Motivated by the discussion above, we choose the gauge function

$$\chi(\vec{\mathbf{r}}, t) = (1/e) \int \vec{\mathbf{A}}(\vec{\mathbf{r}}) \cdot \vec{\mathbf{P}}(\vec{\mathbf{r}}) dV \quad [\text{IV-21}]$$

where  $\vec{\mathbf{P}}(\vec{\mathbf{r}})$  is the polarization density in the form shown in Equation [ IV-19 ] and  $\vec{\mathbf{A}}(\vec{\mathbf{r}})$  is the vector potential appearing in Equation [ IV-14 ]. The impact of the transformation on the scalar potential term in Equation [ IV-14 ] is easy to evaluate -- viz.

$$\chi(\vec{\mathbf{r}}_i, t) = \chi(\vec{\mathbf{r}}_i, t) - \vec{\mathbf{r}}_i \cdot \frac{\exp(\vec{\mathbf{r}}_i - \vec{\mathbf{r}}) - 1}{\vec{\mathbf{r}}_i} \vec{\mathbf{E}}_T(\vec{\mathbf{r}}) \Big|_{\vec{\mathbf{r}}=0} \quad [\text{IV-22}]$$

Dealing with the transformation

$$\vec{A}(\vec{r}_i, t) = \vec{A}(\vec{r}_i, t) - (1/e) \int_i \vec{A}(\vec{r}) \cdot \vec{P}(\vec{r}) dV \quad [IV-23]$$

is straightforward, but extremely tedious (and not very useful)! Substituting for  $\vec{P}(\vec{r})$  from Equation [IV-19] and doing a lot of integrating by parts we could demonstrate that

$$\vec{A}(\vec{r}_i, t) = -\mu_0 \vec{r}_i \times \frac{\exp(\vec{r}_i \cdot \vec{r}) (\vec{r}_i \cdot \vec{r} - 1) + 1}{(\vec{r}_i \cdot \vec{r})^2} \vec{H}(\vec{r}) \Big|_{\vec{r}=0} \quad [IV-24]$$

Making use of these results, the complete Hamiltonian of a atom plus radiation field may be written

$$\begin{aligned} \mathcal{H} = & \frac{1}{2m} \vec{p}_i^2 - e\mu_0 \vec{r}_i \times \frac{\exp(\vec{r}_i \cdot \vec{r}) (\vec{r}_i \cdot \vec{r} - 1) + 1}{(\vec{r}_i \cdot \vec{r})^2} \vec{H}(\vec{r}) \Big|_{\vec{r}=0} \\ & - \frac{1}{2} e^2 \left( \vec{r}_i \right) + \frac{1}{2} Ze^2 (0) + \frac{1}{2} \int \left[ \epsilon_0 \vec{E}_T^2 + \mu_0 \vec{H}^2 \right] dV \\ & + e \int_i \vec{r}_i \cdot \frac{\exp(\vec{r}_i \cdot \vec{r}) - 1}{\vec{r}_i \cdot \vec{r}} \vec{E}_T(\vec{r}) \Big|_{\vec{r}=0} \end{aligned} \quad [IV-25]$$

In practical terms, the *useful* complete Hamiltonian is written

$$\begin{aligned} \mathcal{H} &= \mathcal{H}_A + \mathcal{H}_R + \mathcal{H}_I \\ &= \mathcal{H}_A + \mathcal{H}_R + \left( \mathcal{H}_{ED} + \mathcal{H}_{EQ} + \mathcal{H}_{MD} + \mathcal{H}_{NL} \right) \end{aligned} \quad [IV-26a]$$

where

$$\mathcal{H}_A = \vec{p}_i^2 / 2m - \frac{1}{2} e^2 \left( \vec{r}_i \right) + \frac{1}{2} Ze^2 (0) \quad [IV-26b]$$

$$\mathcal{H}_R = \frac{1}{2} \int_0 \left[ \vec{\mathbf{E}}_T^2 + \mu_0 \vec{\mathbf{H}}^2 \right] dV \quad [\text{IV-26c}]$$

$$\mathcal{H}_{ED} = e \sum_i \vec{\mathbf{r}}_i \cdot \vec{\mathbf{E}}_T(0) = e \vec{\mathcal{D}} \cdot \vec{\mathbf{E}}_T(0) \quad [\text{IV-26d}]$$

$$\mathcal{H}_{EQ} = \frac{1}{2} e \sum_i \vec{\mathbf{r}}_i \cdot \left\{ \left[ \vec{\mathbf{r}}_i \cdot \nabla \right] \vec{\mathbf{E}}_T(\vec{\mathbf{r}}) \right\}_{\vec{\mathbf{r}}=0} = - \vec{\mathcal{Q}} \cdot \vec{\mathbf{E}}_T(\vec{\mathbf{r}})_{\vec{\mathbf{r}}=0} \quad [\text{IV-26e}]$$

$$\mathcal{H}_{MD} = \frac{e \mu_0}{2m} \sum_i \left[ \vec{\mathbf{r}}_i \times \vec{\mathcal{P}} \right] \cdot \vec{\mathbf{H}}(0) = \frac{e \mu_0}{2m} \vec{\mathcal{M}} \cdot \vec{\mathbf{H}}(0) \quad [\text{IV-26f}]$$

$$\mathcal{H}_{NL} = \frac{e^2 \mu_0^2}{8m} \sum_i \left[ \vec{\mathbf{r}}_i \times \vec{\mathbf{H}}(0) \right]^2 \quad [\text{IV-26g}]$$

If we take  $\vec{\mathbf{r}}_i$  of the order of magnitude of the Bohr radius  $a_B$ ,  $\vec{\mathcal{M}}$  of order  $\hbar$ , and  $\vec{\mathbf{E}}_T(\vec{\mathbf{r}})_{\vec{\mathbf{r}}=0}$  of order  $|\vec{\mathbf{k}}| |\vec{\mathbf{E}}_T(0)|$  then we may estimate the three linear interaction terms<sup>25</sup> -- viz.

$$\mathcal{H}_{ED} \sim \mathbf{E}_T(0) e a_B = \mathbf{E}_T(0) \frac{4}{m e} \hbar^2$$

$$\mathcal{H}_{EQ} \sim \mathbf{E}_T(0) \frac{3 e \hbar}{16 m c}$$

$$\mathcal{H}_{MD} \sim \mathbf{H}(0) \frac{\mu_0 e \hbar}{2 m} = \mathbf{E}_T(0) \frac{e \hbar}{2 m c}$$

Further we note that  $\frac{\mathcal{H}_{EQ}}{\mathcal{H}_{ED}}$  and  $\frac{\mathcal{H}_{MD}}{\mathcal{H}_{ED}} = \frac{e^2}{4 \hbar c} = \frac{1}{137} !!$

<sup>25</sup> We assume here that  $\hbar = \hbar c k = e^2 / 2 a_B = R / 2$